Dimensional crossover of thermal conductance in nanowires

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Abstract

Dimensional dependence of thermal conductance at low temperatures in nanowires is studied using the nonequilibrium Green's function (NEGF) method. Our calculation shows a smooth dimensional crossover of thermal conductance in nanowire from one-dimensional to three-dimensional behavior with the increase of diameters. The results are consistent with the experimental findings that the temperature dependence of thermal conductance at low temperature for diameters from tens to hundreds nanometers will be close to Debye law. The calculation also suggests that universal thermal conductance is only observable in nanowires with small diameters. We also find that the interfacial thermal conductance across Si and Ge nanowire is much lower than the corresponding value in bulk materials.

Thermal properties of semiconductor nanowires have attracted significant attention in recent years with the continuous scaling down of feature sizes in microelectronic devices and circuits.[1,2,3,4] Semiconductor nanowires promise applications in future generation electronic and optoelectronic devices. The reduced dimension effects on thermal transport in nanowires becomes important both for device reliability and for intrinsic physics. For one-dimensional quantum atomic chains[1], thermal conductance is proportional to the temperature T at low temperatures with the quantized universal coefficient $\pi^2 k_B^2/3h$. For three-dimensional bulk materials, it is well known that thermal conductance depends on temperature as Debye T^3 law at low temperatures. With the dimension falling between one and three dimensions, the behavior of thermal transport in quasi-one-dimensional nanowires will be an interesting problem to be investigated. Recent experimental results of thermal conductance[2] in Si nanowires with the diameter 22 nm exhibited a deviation from Debye law. The temperature dependence of thermal conductance[3] in Si nanowires with a cross section of $130 \times 200\,\mathrm{nm}^2$ was shown to behave as T^3 above 1.2 K. It is, therefore, important to systematically explore the dimensional effects on thermal conductance in nanowires.

In this letter, we model thermal transport in nanowires using the nonequilibrium Green's function method (NEGF)[5,6,7,8]. We find a one-dimensional to three-dimensional transition of thermal conductance at low temperatures in nanowires. Interfacial thermal conductance across Si and Ge nanowires is found substantially smaller in comparison with the value across bulk Si and Ge epitaxial interface.

We consider the Si semiconductor nanowires as an example. Unlike the three-dimensional bulk materials, there is no translational invariance in the transverse direction in a nanowire. We choose a conventional supercell that includes all the atoms in the transverse directions. Nanowire structures are first optimized using Tersoff potential[9]. Force constants for each atom are obtained from the equilibrium position under small displacements. We have verified that the force constants from Tersoff potential reproduce reasonably well the phonon dispersion of bulk silicon material[10]. Thermal transport in nanowires is calculated along the [100] direction.

Phonon transport in nanowires is treated using the nonequilibrium Green's function formalism, as described in Ref. 5,6,7,8. Thermal current expression for the lead, for example

the left lead, is given by the formula as

$$I = -\frac{1}{2\pi}\int_{-\infty}^{+\infty} d\omega\, \hbar\omega\, \mathrm{Tr}\Big(G^r[\omega]\Sigma_L^<[\omega] + G^<[\omega]\Sigma_L^a[\omega]\Big), \tag{1}$$

where $G^r[\omega]$ and $G^<[\omega]$ are the retarded and the lesser Green's function for the scattering region, respectively. The subscript L denotes the left lead. The lesser self-energy $\Sigma_L^<$ and the advanced self-energy Σ_L^a account for the coupling of the scattering region with the left lead. Similar expressions can be written down for thermal current on the right lead. The retarded Green's function G^r is obtained from the solution of the Dyson equation, as

$$G^r[\omega] = \Big((\omega + i0^+)^2\mathbf{I} - K_c - \Sigma_R^r - \Sigma_L^r - \Sigma_n^r\Big)^{-1}, \tag{2}$$

where K_c is the dynamic matrix for the central scattering region. Here Σ_L^r, Σ_R^r and Σ_n^r are the retarded self-energies due to the coupling with the left/right lead and from the nonlinear phonon-phonon interaction, respectively. The retarded self-energy for the left/right lead $\Sigma_\alpha^r, (\alpha = L, R)$, is calculated through the relation $\Sigma_\alpha^r = V_\alpha g_\alpha^r V_\alpha^\dagger, \alpha = L, R$. Here g_α^r is the surface Green's function which can be calculated through a recursive iteration method[8]. The matrix V_α is the coupling matrix between the semi-infinite lead and the central region. The nonlinear self-energy Σ_n^r can be computed through the expansion of Feynman diagrams or the mean-field theory[8]. Here we concentrate on the dimensional dependence of thermal transport in nanowires at low temperatures. We will ignore the nonlinear interactions. This approximation is reasonable because the phonon boundary instead of the phonon-phonon scattering dominates thermal transport at moderation high temperatures in nanowires[2]. If the nonlinear self-energy is not considered, Eq. (1) can be further reduced to the Landauer formula[6,7,8]. The thermal conductance $G = \frac{1}{S_0}\partial I/\partial T$,where S_0 is the cross-section of the nanowire, in terms of Landauer formula is given as

$$G = \frac{1}{2\pi\, S_0}\int_0^\infty d\omega\, \hbar\omega\, \mathcal{T}[\omega]\frac{\partial f}{\partial T}, \tag{3}$$

where the Caroli transmission is $\mathcal{T}[\omega] = \mathrm{Tr}(G^r\Gamma_L G^a\Gamma_R)$ and $\Gamma_\alpha = i(\Sigma_\alpha^r - \Sigma_\alpha^a), \alpha = L, R$.

The phonon dispersion relation for Si nanowire with the diameter $d = 1.54\,\mathrm{nm}$ is illustrated in Fig. 1. It can be seen from Fig. 1 that there are three acoustic branches: one longitudinal branch and two degenerately transverse branches. The maximum frequency for acoustic branches in Si nanowire is $36\,\mathrm{cm}^{-1}$, which is in contrast with the value of about $340\,\mathrm{cm}^{-1}$ for Si bulk[10]. It can be seen for Fig. 1 that many optical branches with small group velocities emerge.

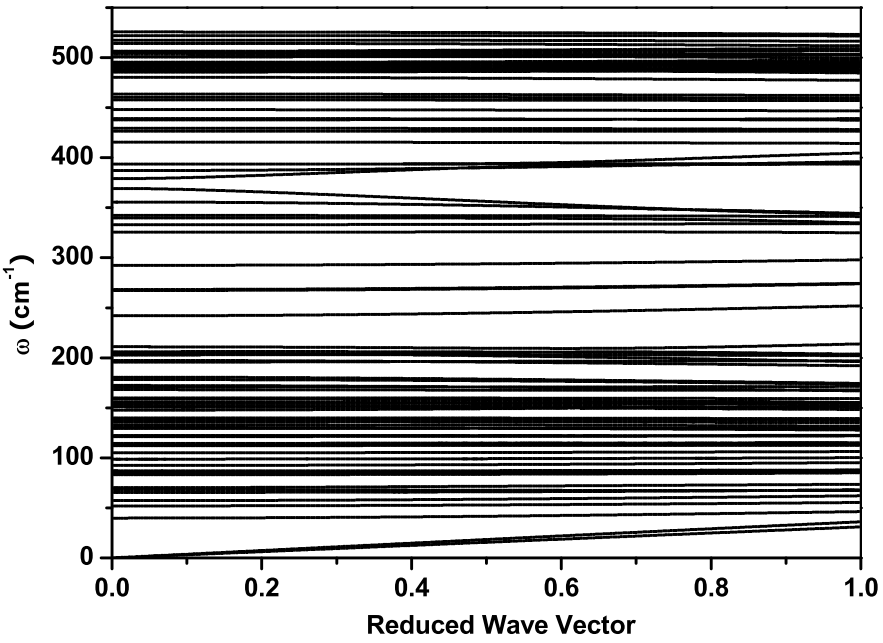

FIG. 1: Phonon dispersions of Si nanowire with the diameter $d = 1.54\,\mathrm{nm}$. The force constants are calculated from Tersoff potential. The wave vectors are in terms of the reduced wave number of the first Brillouin zone.

Thermal conductance calculated from NEGF for Si nanowires with the diameters ranging from 1.54 nm to 6.14 nm is shown in Fig. 2. For comparison, thermal conductance for bulk silicon calculated with the force constants derived from Tersoff potential is also plotted in Fig. 2. When temperature $T > 60K$, thermal conductance increases with the diameters of Si nanowires, but it is still below the value of bulk Si material. It can be explained by the fact that the optical branches which dominate thermal conductance in Si nanowires have smaller group velocities. This kind of diameter-dependent behavior is also consistent with the experimental results in Ref. 2. The most significant feature in Fig. 2 is the temperature dependence of thermal conductance with the increase of the diameter of nanowires at low temperatures. The temperature dependence of the thermal conductance $G \propto T^{\alpha}$ below 60K is plotted on a log-log scale in Fig. 2(B). It can be seen from Fig. 2(B) that the exponent α changes from $\alpha = 1.3$ to $\alpha = 2.6$ with the increase of diameters. This dimensional crossover of thermal conductance from one-dimensional T behavior to three-dimensional Debye T^3 law is clearly seen in Fig. 2(B). It can be concluded from this figure that universal thermal conductance can only be observed in nanowires with very small diameters.

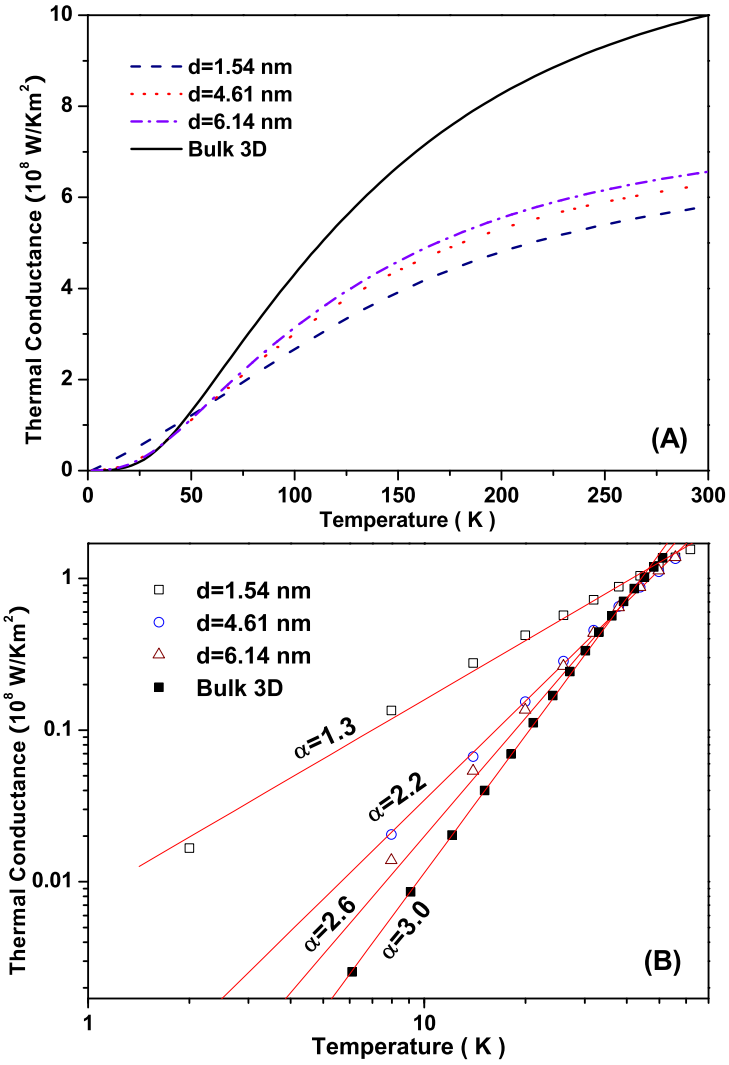

FIG. 2: **(A)** Thermal conductance for Si nanowires with different diameters and for Si bulk. **(B)** The log-log plot of thermal conductance at low temperatures.

To understand the above dimensional crossover behavior, we plot the transmission $\mathcal{T}[\omega]$ dependence on frequency ω for Si nanowires with different diameters in Fig. 3. We can assume that the transmission $\mathcal{T}$ has a dependence on frequency with the relation $\mathcal{T} \propto \omega^{\beta}$ in the range of low frequencies. For one dimension, the transmission equals 1 so that $\beta = 0$. For three-dimensional bulk, ballistic thermal conductance G_{bulk} at low temperature can be written as $G_{bulk} = \frac{1}{(2\pi)^3}\Sigma_s \int d^3\mathbf{q}\, \hbar\omega_s\, v_s^z \frac{\partial f}{\partial T}$, where s denotes the different polarized branch. Compared with Eq. (3), the transmission $\mathcal{T}_{bulk}[\omega]$ for bulk material is given as

$$\mathcal{T}_{bulk}[\omega] = \frac{1}{(2\pi)^2}\Sigma_s \int d^3\mathbf{q}\, \delta(\omega - \omega_s(\mathbf{q}))v_s^z. \tag{4}$$

Note that $\mathcal{T}_{bulk}[\omega]$ in Eq.(4) is equivalent to $\mathcal{T}[\omega]/S_0$ in Eq.(3). In the low-frequency range,

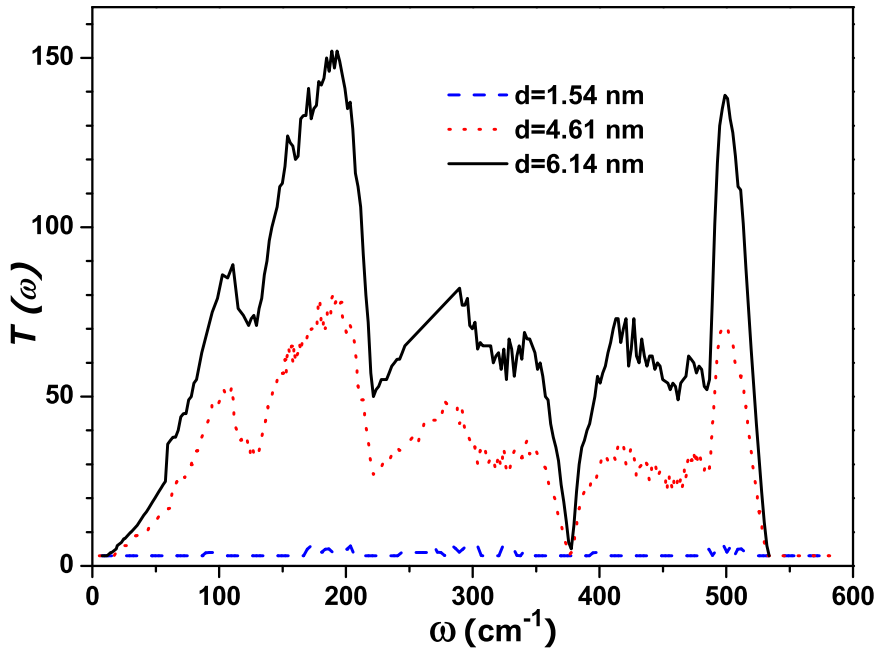

FIG. 3: The dependence of transmission function $\mathcal{T}$ on frequencies for Si nanowires with different diameters.

only the acoustic branches need to be considered for three-dimensional bulk materials. If we use the Debye model, then all three branches of the spectrum have the linear dispersion relation as $\omega_s = c_s q$. Substituting this relation into Eq.(4), we find that the transmission for bulk material is given as $\mathcal{T}_{bulk}[\omega] = \Sigma_s(\frac{\omega}{c_s})^2/4\pi \propto \omega^2$. Thus the transmission function at low frequency for three dimensional-bulk depends quadratically on the frequency, i.e. $\beta = 2$. It is a straightforward conclusion from Eq.(3) that thermal conductance G at low temperature will behave as $T^{\beta+1}$, *i.e.* $\alpha = \beta + 1$, if the transmission function depends on frequency as T^{β}. It can be seen from Fig. 3 that the exponent β for the transmission at low frequency increases with the diameters of nanowires. This explains why the temperature dependence of thermal conductance at low temperature increases from $\alpha = 1.3$ to $\alpha = 2.6$ with the diameters from $d = 1.54\,\mathrm{nm}$ to $d = 6.14\,\mathrm{nm}$.

Furthermore we investigate interfacial thermal conductance in nanowires using NEGF. Thermal conductance in the Si and Ge superlattice nanowires[4] was found very small in comparison with the value of bulk materials. Our aim is to understand the role played by the Si-Ge interface in nanowires. The force constants for Si and Ge used in the calculation are also derived from Tersoff potential after the optimization of structures. The thermal

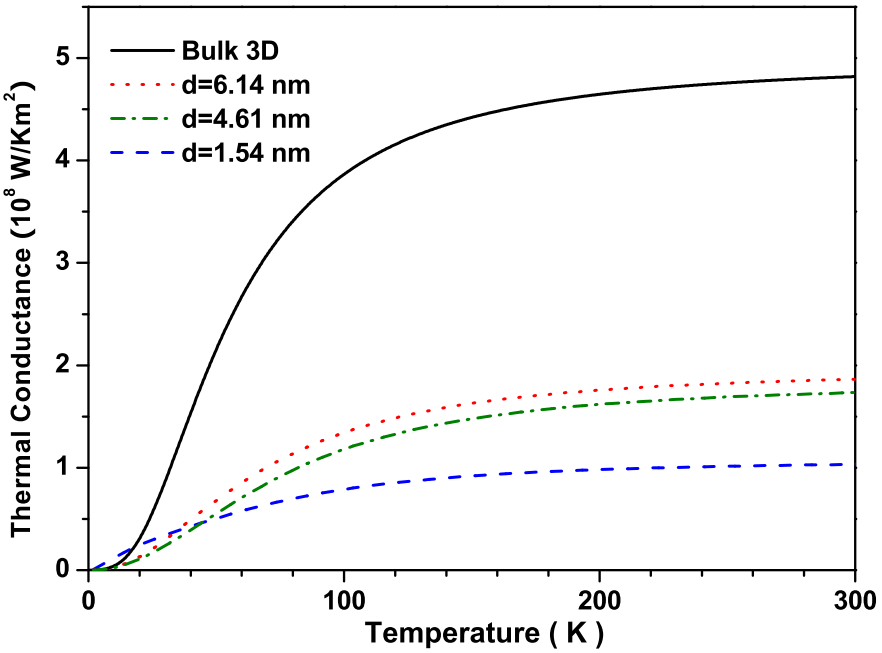

FIG. 4: Temperature dependence of interfacial thermal conductance for Si-Ge nanowires with different diameters d = 1.54, 4.61, 6.14 nm and for the epitaxial interface between bulk Si and Ge (the solid line).

conductance across silicon and germanium nanowires with different diameters is plotted in Fig. 4. For comparison, epitaxially interfacial thermal conductance between bulk silicon and germanium is calculated using the mode-matching lattice dynamic method[11,12]. It can be seen from Fig.4 that interfacial thermal conductance across Si and Ge nanowires shows a similar dimensional crossover of temperature dependence like that of pure Si nanowires at temperature below 60K. When the temperature is above 60K, interfacial thermal conductance increase with the diameters of nanowires. Thermal conductance at temperature T = 200K between Si and Ge nanowires with the diameter d = 6.14nm is 1.7×10^8W/Km^2, which is about one third of thermal conductance 4.6×10^8W/Km^2 at the same temperature between bulk Si and Ge epitaxial interface. In contrast with thermal conductance 5.6×10^8W/Km^2 in pure Si nanowire with the same diameter, thermal conductance across Si-Ge nanowires is about one fourth of it. This substantially decreased thermal conductance across Si and Ge nanowires may result from the reduced group velocity for optical branches in nanowires.

In summary, thermal conductance in nanowires is calculated using NEGF. Our calcula-

tion shows a clear dimensional crossover for temperature dependence of thermal conductance in nanowires at low temperatures. We conclude that thermal conductance at low temperature in most experimental nanowires with diameters $d > 10\,\mathrm{nm}$ will behave close to T^3. At moderately high temperature, thermal conductance will increase with the nanowire diameter and tends to the upper limit for the corresponding bulk material. The small value of interfacial thermal conductance across Si-Ge nanowires is one of the reasons that account for substantial reduction of thermal conduction in superlattice nanowires. Our present calculation holds at low, or moderate, temperature when phonon-phonon scattering does not play a dominant role in thermal conductance in low-dimensional materials. When temperature is high enough, nonlinear phonon-phonon scattering should be included.

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